

Citation for published version:

Coleman, P, Culver, NJ & Dowler, BMW 2013, 'Positronium formation in methanol and ethanol', *Physical Review A: Atomic, Molecular, and Optical Physics*, vol. 87, no. 1, 012712. <https://doi.org/10.1103/PhysRevA.87.012712>

DOI:

[10.1103/PhysRevA.87.012712](https://doi.org/10.1103/PhysRevA.87.012712)

Publication date:

2013

Document Version

Publisher's PDF, also known as Version of record

[Link to publication](#)

N J Culver, & B M W M Dowler (2013). Positronium formation in methanol and ethanol. *Physical Review A - Atomic, Molecular, and Optical Physics*, 87(1), 012712.

Copyright 2013 by the American Physical Society

University of Bath

Alternative formats

If you require this document in an alternative format, please contact:
openaccess@bath.ac.uk

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Positronium formation in methanol and ethanol

P. G. Coleman, N. J. Culver, and B. M. W. M. Dowler

Department of Physics, University of Bath, Bath BA2 7AY, United Kingdom

(Received 18 December 2012; published 31 January 2013)

Cross sections for the formation of positronium (Ps) in collisions of positrons with methanol and ethanol molecules, Q_{Ps} , have been measured for positrons of incident energies from 3 to 40 eV. The threshold energies for Ps formation were found to be 4.0 ± 0.2 eV and 3.5 ± 0.5 eV, respectively. Q_{Ps} for the two molecules are very similar in size and energy dependence, suggesting that Ps formation is associated preferentially with the OH group, whereas $(Q_{\text{tot}} - Q_{\text{Ps}})$, where Q_{tot} is the total scattering cross section, is related more to the physical size of the molecules.

DOI: [10.1103/PhysRevA.87.012712](https://doi.org/10.1103/PhysRevA.87.012712)

PACS number(s): 34.80.Uv, 36.10.Dr

I. INTRODUCTION

Over the past decade there has been increasing interest in the total scattering cross sections (Q_{tot}) for positrons interacting with biomolecules (e.g., the recent series of measurements by Zecca and co-workers presented in Refs. [1–5]). The motivation for these measurements is to advance the understanding of the interaction of positrons in matters of biological importance, for example, in improving the quality of modeling of positron tracks in positron emission tomography (PET). Of particular interest in this last example is the probability of positronium (Ps) formation in tissue, which can be significant and thus affect tomographic interpretation. In one of the early papers in their biomolecule series Zecca *et al.* [1] recognize the potential importance of the Ps formation probability and point to discontinuities in their measured Q_{tot} for methanol and ethanol at positron energies close to the expected thresholds E_{Ps} for Ps formation. $E_{\text{Ps}} = (E_i - 6.8)$ eV, where E_i is the first ionization potential and 6.8 eV is the ground-state binding energy of Ps, giving $E_{\text{th}} = 4.0$ and 3.7 eV for methanol and ethanol, respectively, both with uncertainties of ± 0.1 eV. However, it is difficult to separate the Ps formation cross section Q_{Ps} from Q_{tot} because the elastic scattering cross section decreases strongly with incident positron energy (E) through E_{th} , and extrapolation is unreliable.

In 2009 Makochekanwa *et al.* [6] reported their measurements of Q_{tot} and Q_{Ps} in water and formic acid using a positron loss technique. In the present paper a version of this technique is used to make direct measurements of Q_{Ps} for methanol and ethanol. These two primary alcohols were chosen as simple organic targets—early steps towards a future wider study of Ps formation in biomolecules and to enable comparison with the Q_{tot} values of Zecca *et al.* [1].

II. EXPERIMENTAL DETAILS

The apparatus used for the current measurements is described by Thornton and Coleman [7]. Positrons from a ^{22}Na source are moderated by two annealed tungsten meshes, and the energy spread of the resulting positron beam is reduced to below 1 eV by applying a cutoff potential to a 92%-transmission mesh held in front of the moderator. This cutoff mesh is held at a constant $(V_{\text{mod}} + 1.6)$ V, where V_{mod} is the moderator potential. The mesh also serves as an effective positron reflector, returning to the forward direction positrons

which are scattered into the backward hemisphere; this is important in the measurement of Q_{Ps} , as discussed below. The positrons, guided by a 7-mT axial magnetic field, pass through a 70-mm-long gas cell and a cylindrical retarding field analyzer (RFA) and are detected by a single-channel electron multiplier (CEM) whose front end is held at -2.0 kV to reduce the background count rate due to energetic secondary electrons from the moderator meshes. The mean positron energy E was found, by scanning the RFA potential, to be $(V_{\text{mod}} + 2)$ eV to within 0.1 eV.

The method is based on measuring positron loss from a 4-mm-diameter beam passing through a gas cell under the influence of the high magnetic field; all scattered positrons surviving a collision are constrained by the field to paths which end on a channel electron multiplier (CEM) detector with a 10-mm-diameter acceptance cone. Those which do not are considered to have formed Ps or to have been annihilated. As discussed by Jones *et al.* [8], enhanced annihilation can occur with molecules like methanol and ethanol below ~ 1 eV incident energy, but the resultant annihilation cross section is still orders of magnitude lower than the scattering cross sections of interest here, and thus direct annihilation as a loss mechanism is neglected. Another problem with these alcohols is the enormous increase in Q_{tot} below a few eV, attributed by Zecca *et al.* [1] to the polarizability of the molecules. The very large Q_{tot} values mean that the thin target approximation breaks down at these low energies, in that double or even multiple scattering can occur, and so here we present results for positron energies of 3 eV and above. At these energies the only significant loss process is thus Ps formation, in which the positron is essentially neutralized and is no longer constrained by the magnetic field. The beam attenuation under these conditions is A_{Ps} .

Using the same apparatus the total attenuation A_{tot} of the beam is measured (with all other conditions unchanged) by discriminating against scattered positrons using the RFA between the gas cell and the CEM which is set to a potential which ideally lets only unscattered positrons pass, i.e., those which have lost no axial momentum. This was found to be $(V_{\text{mod}} + 1.6)$ V, where V_{mod} is the potential applied to the positron moderator.

In a guiding magnetic field of 7 mT this RFA method for measuring A_{tot} is reliable only when the fraction of positrons elastically scattered at angles below the minimum differentiable by the technique is negligible. In the current

measurements, in which the full width at half maximum of the beam's energy spectrum was a little under 1 eV, this was found to be true for $E > 15$ eV, when the measured A_{tot} led to Q_{tot} values which agreed extremely well up to 40 eV with those of Zecca *et al.* [1] by choosing an appropriate (single) value for the gas density-path length product nL in the Beer-Lambert expression:

$$Q_{\text{tot}} = -[\ln(1 - A_{\text{tot}})]/nL. \quad (1)$$

Below 15 eV, this derived value for nL was used to compute A_{tot} from Eq. (1) using the Q_{tot} values of Zecca *et al.* [1]. It is interesting to note that the only other measurements for positron Q_{tot} in methanol or ethanol, those by Kimura *et al.* [9] for methanol, also only agree with Zecca *et al.* [1] above ~ 15 eV, probably for the same reason.

The Ps formation cross section at any incident positron energy, for a thin target, is then

$$Q_{\text{Ps}} = (A_{\text{Ps}}/A_{\text{tot}})Q_{\text{tot}}, \quad (2)$$

where the Q_{tot} values from Ref. [1] are used.

To approximate to the thin-target (single scattering) condition, multiple scattering effects were minimized by limiting nL , thereby restricting A_{tot} to a maximum of $\sim 20\%$. The RFA was held at a measured -0.25 V, rather than at 0.00 V, during the A_{Ps} measurements to avoid any contact potential effects; this was found to make a small but measurable difference at low incident positron energies.

Total and background CEM count rates at each positron energy E (in ~ 1 eV steps between 2 and 40 eV) were collected with and without gas in the gas cell, and with and without ($V_{\text{mod}} + 1.6$) V applied to the RFA, i.e., eight measurements at each energy E . The background rates were measured by raising the potential on the fine cutoff mesh to 3.5 V above V_{mod} . In order to avoid any drift-related errors, data were collected using a multichannel scaler, by which CEM counts were recorded by combining 200 scans from 2 to 40 eV, each individual dwell time being 10 s. Without gas in the cell the dependencies of the count rates on E were smoothly varying and small, so that fits of the data could be used to decrease statistical uncertainties, as could the background data in gas. The most significant runs, i.e., those used in the determination of A_{Ps} , were repeated and/or extended to improve statistical precision.

It was important to counter any effects of drifts in system performance *between* these relatively long runs. To do this count rates under all eight conditions described above, in vacuum and in gas, were measured at selected values of E within minutes of each other; the relative count rates thus measured were then used to normalize all the long runs.

III. RESULTS AND DISCUSSION

The results for Q_{Ps} in methanol and ethanol are presented in Figs. 1 and 2. The scatter in the points indicates statistical precision. There are presently no other measurements or calculations with which these results can be compared; however, in Figs. 1 and 2 the results have been subtracted from the Q_{tot} of Zecca *et al.* [1] and demonstrate that the magnitude of Q_{Ps} appears to fit well into that of the total cross section. The threshold energies suggested by the data for Ps formation in methanol and ethanol are 4.0 ± 0.2 eV and 3.5 ± 0.5 eV,

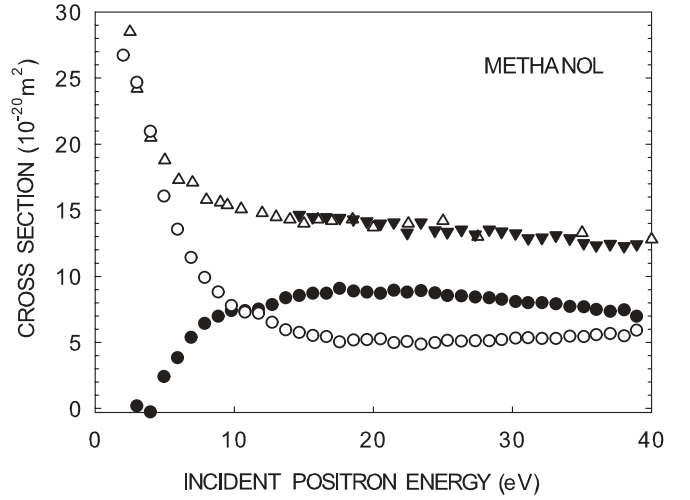


FIG. 1. Cross sections for positron-methanol interactions. Solid circles: Q_{Ps} . Inverted solid triangles: Scaled Q_{tot} . Open triangles: Q_{tot} of Zecca *et al.* [1] Open circles: Q_{tot} (Zecca *et al.* [1]) $- Q_{\text{Ps}}$.

respectively, both consistent with the expectations outlined in Sec. I above.

It is evident that Q_{Ps} is a significant fraction of Q_{tot} in the energy range from threshold to 40 eV, contributing ~ 65 and 50% of the scattering in methanol and ethanol at its peak. The somewhat larger total cross sections for ethanol are attributed in the review by Brunger *et al.* [10] to its higher dipole polarizability, rather than its larger physical size. However, we note that Q_{Ps} values for both alcohols are very similar in magnitude (to within $\sim 10\%$) and energy dependence, whereas $(Q_{\text{tot}} - Q_{\text{Ps}})$ for ethanol is approximately double that for methanol. The data therefore suggest the following: (i) the molecular dipole polarizabilities have a strong effect at low (< 5 eV) positron energies, where Q_{tot} increases dramatically; (ii) above ~ 5 eV the polarizabilities have a much smaller effect on the cross sections and the larger $(Q_{\text{tot}} - Q_{\text{Ps}})$ for ethanol—dominated by the elastic scattering cross section over most of the energy range studied here—is a consequence of its larger size; and (iii) the almost identical $Q_{\text{Ps}}(E)$ for methanol

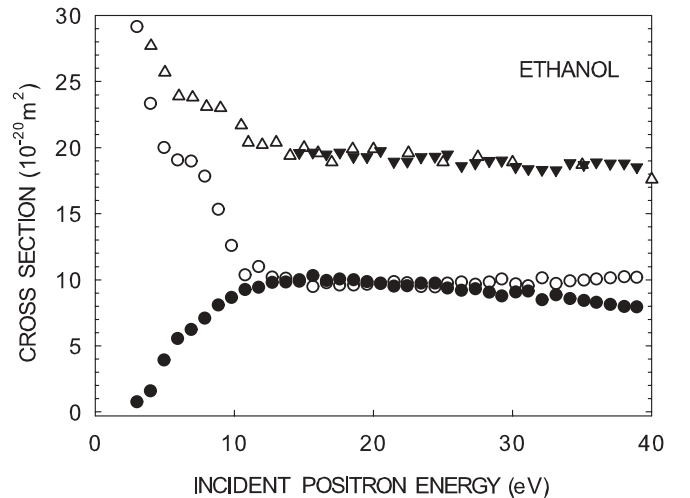


FIG. 2. Cross sections for positron-ethanol interactions. Symbols are as defined in Fig. 1.

and ethanol suggests that Ps formation is associated primarily with the OH group. Postulate (iii) implies that the saturated C-H structures attached to the OH group do not make a significant contribution to Ps formation and conversely that the electronegativity of oxygen and the electronic structure of the OH group dominate this process. It is difficult to separate the elastic contributions from the inelastic contributions to ($Q_{\text{tot}} - Q_{\text{Ps}}$) but the small, comparable increase at higher energies is probably associated with ionization, which might also be linked to the OH group and thus be similar in the two alcohols.

IV. CONCLUSIONS

The present results for methanol and ethanol are similar in energy dependence and magnitude, are similar to the only other

published Q_{Ps} for an organic molecule (formic acid) [6], and suggest that, for simple organic molecules at least, an almost universal form can be adopted in work in which $Q_{\text{Ps}}(E)$ is required.

Our results support the conclusions of Zecca and co-workers [1–5] that Ps formation and decay is an important process in biomolecular systems, and their understanding is important if reliable modeling of positron tracks in PET is to be achieved.

ACKNOWLEDGMENTS

The authors are grateful to Harry Bone, Charlene Edwardson, and Sam Townrow for help and encouragement.

-
- [1] A. Zecca, L. Chiari, A. Sarkar, K. L. Nixon, and M. J. Brunger, *Phys. Rev. A* **78**, 022703 (2008).
 - [2] A. Zecca, L. Chiari, A. Sarkar, M. A. P. Lima, M. H. F. Bettega, K. L. Nixon, and M. J. Brunger, *Phys. Rev. A* **78**, 042707 (2008).
 - [3] A. Zecca, L. Chiari, K. L. Nixon, M. J. Brunger, S. Chattopadhyay, D. Sanyal, and M. Chakrabarti, *J. Phys. Chem. A* **113**, 14251 (2009).
 - [4] A. Zecca, L. Chiari, G. Garcia, F. Blanco, E. Trainotti, and M. J. Brunger, *J. Phys. B* **43**, 215204 (2010).
 - [5] L. Chiari, A. Zecca, S. Girardi, A. Defant, F. Wang, X. G. Ma, M. V. Perkins, and M. J. Brunger, *Phys. Rev. A* **85**, 052711 (2012).
 - [6] C. Makochekanwa, A. Bankovic, W. Tattersall, A. Jones, P. Caradonna, D. S. Slaughter, K. Nixon, M. J. Brunger, Z. Petrovic, J. P. Sullivan, and S. J. Buckman, *New J. Phys.* **11**, 103036 (2009).
 - [7] M. J. Thornton and P. G. Coleman, *J. Phys. B* **44**, 145201 (2011).
 - [8] A. C. L. Jones, J. R. Danielson, J. J. Gosselin, M. R. Natisin, and C. M. Surko, *New J. Phys.* **14**, 015006 (2012).
 - [9] M. Kimura, O. Sueoka, A. Hamada, and Y. Itikawa, *Adv. Chem. Phys.* **111**, 537 (2000).
 - [10] M. J. Brunger, S. J. Buckman, and A. Zecca, *J. Phys.: Conf. Ser.* **194**, 012034 (2009).